

This Page Is Inserted by IFW Operations
and is not a part of the Official Record

BEST AVAILABLE IMAGES

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images may include (but are not limited to):

- BLACK BORDERS
- TEXT CUT OFF AT TOP, BOTTOM OR SIDES
- FADED TEXT
- ILLEGIBLE TEXT
- SKEWED/SLANTED IMAGES
- COLORED PHOTOS
- BLACK OR VERY BLACK AND WHITE DARK PHOTOS
- GRAY SCALE DOCUMENTS

IMAGES ARE BEST AVAILABLE COPY.

**As rescanning documents *will not* correct images,
please do not report the images to the
Image Problem Mailbox.**



(12) UK Patent (19) GB (11) 2 037 726 B

SCIENCE REFERENCE LIBRARY

(54) Title of invention

A process for the pyrogenic production of very finely divided oxides or a metal and/or metalloid

(51) INT CL³; C01B 33/12

(21) Application No
7939469

(22) Date of filing
14 Nov 1979

(30) Priority data

(31) 2849851

(32) 17 Nov 1978

(33) Fed. Rep of Germany (DE)

(43) Application published
16 Jul 1980

(45) Patent published
3 Nov 1982

(52) Domestic classification
C1A 13 N34 N4 N54 PB4

(56) Documents cited
EP (GB) 0011084

(58) Field of search
C1A

(73) Proprietor
Deutsche Gold und Silber
Scheideanstalt Vormal's Roessler,
Weissfrauenstrasse 9,
6 Frankfurt Main,
Germany.

(72) Inventors
Ludwig Lange,
Josef Schmid,
Hans Klebe Dieter Schutte.

(74) Agents
Elkington & Fife,
High Holborn House,
52/54 High Holborn,
London WC1V 6SH.

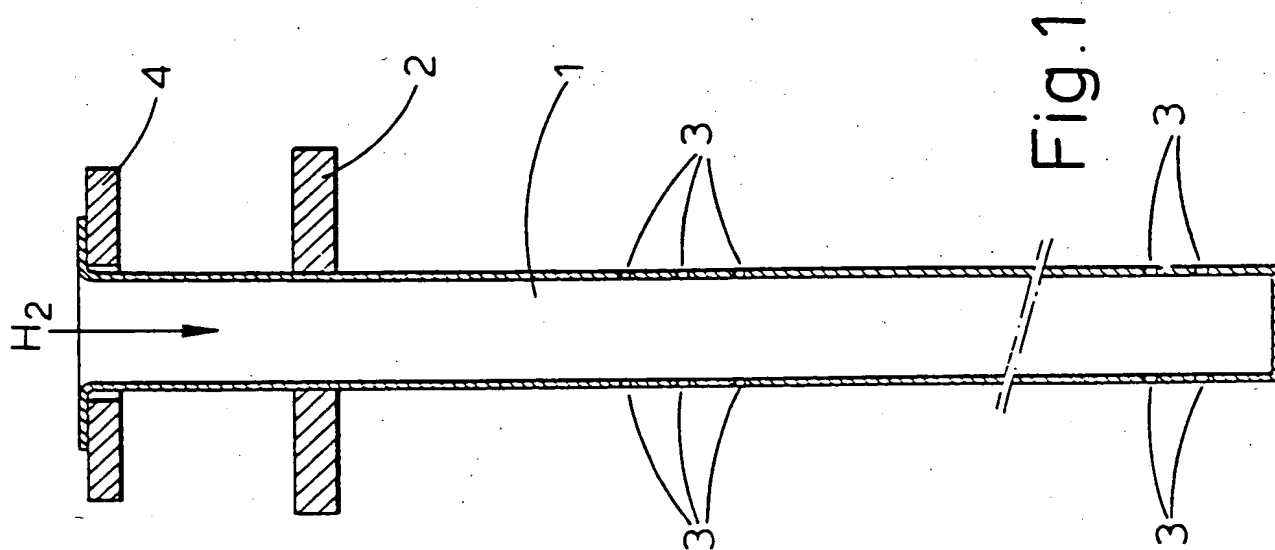


Fig. 1

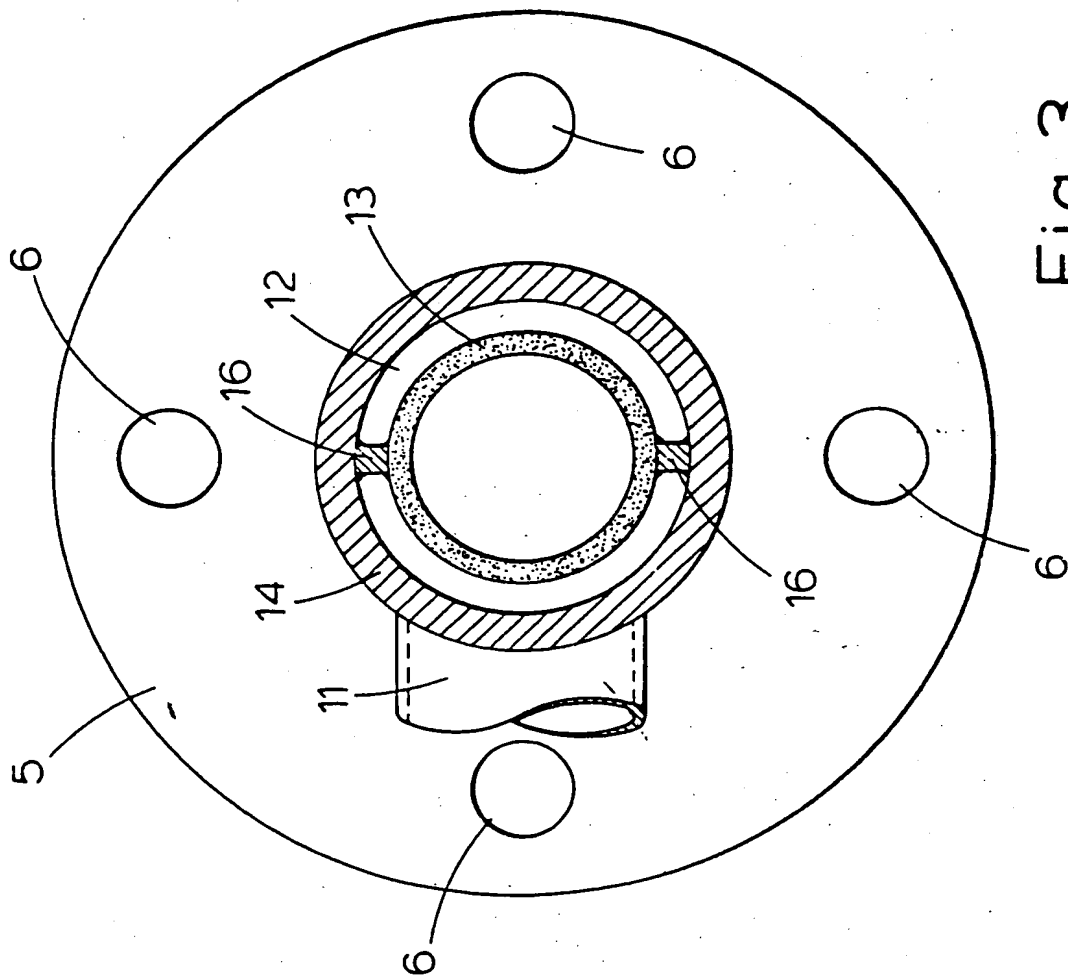
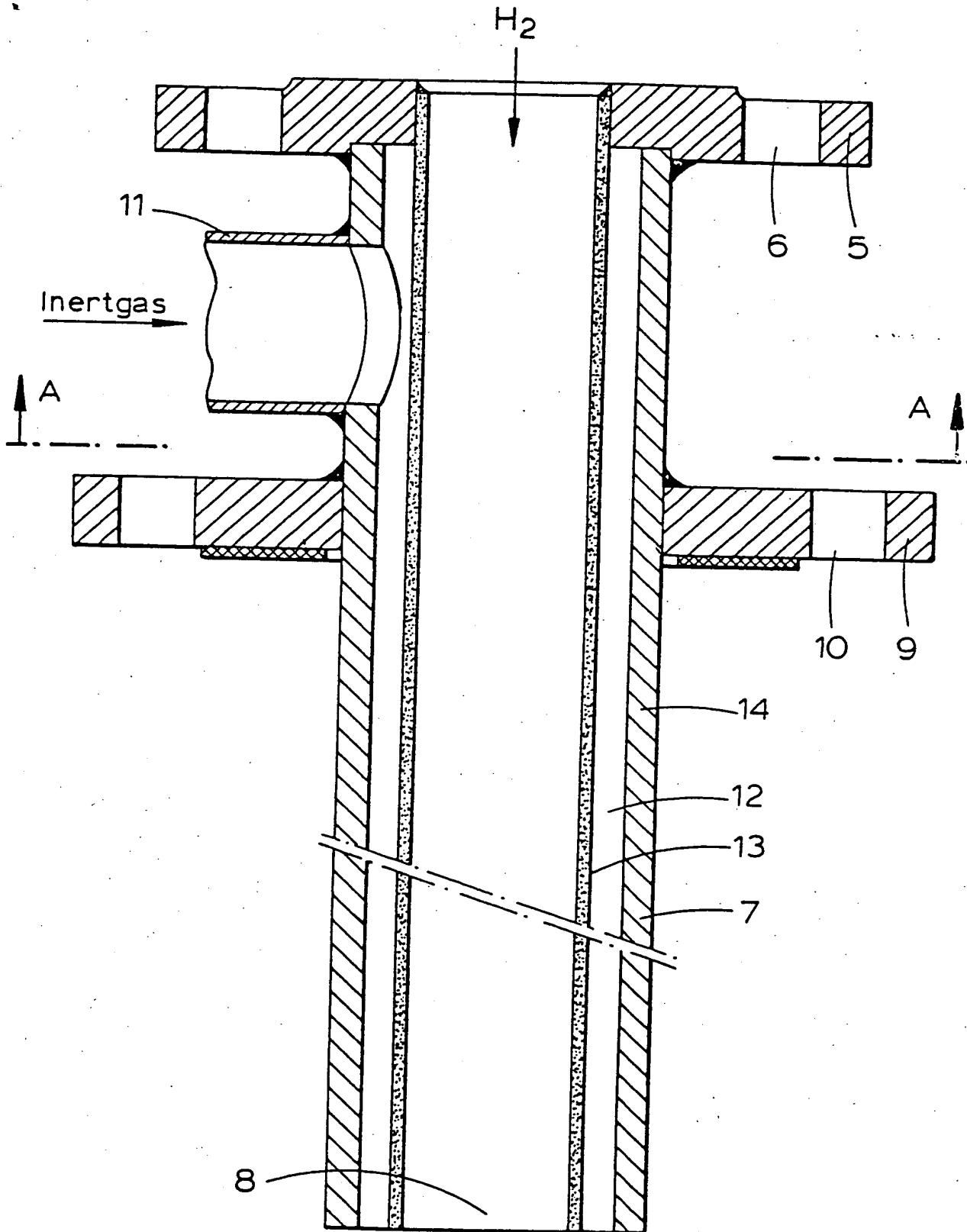


Fig. 3



SPECIFICATION

A process for the pyrogenic production of very finely divided oxides of a metal and/or metalloid

5 The present invention relates to a process for the pyrogenic production of very finely divided oxides of a metal and/or metalloid. More particularly, it relates to a process for the pyrogenic production of a very
10 finely divided oxide of a metal and/or metalloid in which a volatile halogen compound of the metal and/or metalloid is used as a starting material and in which the elementary halogen formed as a secondary product during formation of the oxide of
15 the metal and/or metalloid is reacted during cooling of the reaction products with additionally introduced gaseous hydrogen below the reaction temperature of the oxyhydrogen gas mixture, consisting of the gaseous hydrogen additionally introduced and the
20 oxygen present in the reaction gases, to form hydrogen halide.

In the pyrogenic production of very finely divided oxides of a metal and/or metalloid, volatile compounds of the metal and/or metalloid, together
25 with inflammable or hydrogen-forming gases and oxygen or oxygen-containing gases (referred to herein as an oxyhydrogen gas mixture), are delivered to a burner either separately or in admixture. The inflammable gas and the
30 oxygen-containing gas are delivered in a quantitative ratio which guarantees both complete combustion of the inflammable gas and also hydrolysis of the volatile compound of the metal and/or metalloid.

35 When inorganic or organic halogen compounds of the metal and/or metalloid are used as starting materials, the metal oxide and/or metalloid oxides accumulate together with a waste gas containing hydrogen halide which is separated from the metal
40 and/or metalloid oxide in corresponding separators. Elementary halogen is formed in a secondary reaction. Depending on the reaction conditions used for the production of the metal oxide and/or metalloid oxide, the quantity of elementary halogen
45 formed amounts to from 6 to 10% by weight, based on the quantity of hydrogen halide formed.

In one known pyrogenic process for the production of oxides of a metal and/or metalloid, the elementary halogen formed, for example chlorine, is
50 removed from the waste reaction gas by reducing the elementary halogen formed with hydrogen during cooling of the reaction products below the reaction temperature of the hydrogen with the oxygen present in the waste reaction gas (German
55 Offenlegungsschrift No. 2,533,925).

In this known process, in which SiCl_4 (silicon tetrachloride) is used as a starting material for the production of silicon dioxide, the mixture formed by the waste reaction gas and silicon dioxide
60 undergoes a reduction in temperature of from 1000 to 200°C during cooling in a relatively long cooling zone. The additional hydrogen is introduced in a region of the cooling zone in which the waste reaction gases still have a temperature of from 500

temperature above 700°C because, at temperatures of this order, the hydrogen reacts with the oxygen. It is also inadvisable to add the hydrogen at a temperature below 500°C because, in this case, the
70 velocity of the reaction of the hydrogen with the elementary chlorine is too slow.

The best results are obtained by introducing the elementary hydrogen at a temperature of from 550 to 630°C.

75 The exact point at which the elementary hydrogen is introduced into the cooling zone is dependent on the load. In other words, in the event of production-induced changes in the rate of flow, the point at which the elementary hydrogen is
80 introduced into the cooling zone has to be changed.

The hydrogen is introduced through a pipe of which the length corresponds to the diameter of the cooling zone. The wall of this pipe comprises two rows of bores through which the hydrogen is
85 introduced into the waste reaction gases. One such pipe is shown in cross-section in Figure 1.

Unfortunately, the known process is attended by the disadvantage that the point at which the hydrogen is introduced has to be changed even in
90 the event of minor changes in load. Another disadvantage lies in the fact that, in some cases, the hydrogen reacts so violently with the elementary chlorine that flames are formed. Accordingly, the reaction becomes uncontrollable and, in some
95 cases, can result in damage to the silicon dioxide present in the waste reaction gas. This involves the danger of premature wear of the pipes through which the hydrogen is introduced.

The present invention relates to a process for the
100 pyrogenic production of very finely divided oxides of a metal and/or metalloid by reacting a volatile halogen compound of the metal and/or metalloid with an oxyhydrogen gas mixture in which elementary halogen formed during formation of the
105 oxide of the metal and/or metalloid is reacted during cooling of the reaction products in a cooling zone with additionally introduced gaseous hydrogen at a temperature below the reaction temperature of the gaseous hydrogen additionally introduced and the
110 oxygen present in the waste reaction gases, to form hydrogen halide, the gaseous hydrogen being introduced into the cooling zone by means of at least one double-walled pipe, and an inert gas being additionally introduced by means of the same
115 double-walled pipe between the outer and inner walls of the said double-walled pipe.

The quantity of additional hydrogen is determined by the reaction conditions for the specific metal oxide or metalloid oxide to be produced. Hydrogen
120 may be introduced in such a quantity that the waste gas ultimately obtained contains from 1.6 to 2.0% by volume of hydrogen.

The hydrogen may be added over the entire cooling zone at several points, for example at from 3
125 to 6 points, a double-walled pipe being inserted at each point.

In one preferred embodiment of the present invention, the elementary hydrogen is introduced into the cooling zone at a temperature of from 500 to

550 to 630°C.

The inert gas used may be a noble gas or carbon dioxide, although nitrogen is particularly preferred. It may be used in a quantity of from 20 to 200 m³/h.

5 Volatile halogen compounds of the metals aluminium or titanium or of the metalloids (semi-metals) silicon or germanium may be used as starting materials for the production of the metal oxides or metalloid oxides.

10 For producing mixed oxides of different elements, a mixture of the volatile halogen compounds of the corresponding elements may be used as the starting material.

In one preferred embodiment, the starting halogen compound used is the corresponding chloride of the metals or metalloids. Organic halogen compounds may also, however, be used. Thus, SiHCl₃, SiCl₂H₂, SiCl₄, CH₃SiCl₃, (CH₃)₂SiCl₂, (CH₃)₃SiCl, CH₃-CH₂-SiCl₃ or (CH₃-CH₂)₂SiCl₂ may be used for the production of silicon dioxide.

The process according to the present invention has the advantage that the hydrogen may be mixed more intensively with the waste reaction gas. By simultaneously adding the inert gas, it is possible to influence the temperature at the point of introduction so that, in the event of changes in the rate of flow, there is no need for the point of introduction to be changed. Thus, the hydrogen may be introduced at a point at which the temperature of the waste reaction gases is so high that the hydrogen would be immediately burnt off with the halogen without this undesirable burning taking place. Another advantage is that, by introducing the inert gas, the pipe through which the hydrogen is introduced is cooled and, hence, remains serviceable for a longer period and the danger of corrosion can be avoided. In addition, the introduction of inert gas prevents metal oxide or metalloid oxide from being deposited on the pipe.

40 The process according to the present invention is described in more detail in the following with reference to the accompanying drawings, wherein:

Figure 1 shows a conventional pipe for the introduction of hydrogen.

45 *Figure 2* is a longitudinal section through the double-walled pipe according to the present invention for the introduction of hydrogen.

Figure 3 is a cross section on the line A - A of the double-walled pipe for the introduction of hydrogen shown in *Figure 2*.

50 As shown in *Figure 1*, the pipe 1 through which the hydrogen is introduced is fixed in the wall of the cooling zone 2 in such a way that its length corresponds to the diameter of the cooling zone. The pipe 1 comprises two rows of bores 3 in its wall. The hydrogen is introduced into the pipe 1 through a line fixed to the flange 4 and flows through the bore 3 into the cooling zone.

As shown in *Figure 2*, the hydrogen is introduced into the double-walled pipe 7 through a line fixed to the flange 5 comprising the bore 6 and emerges at the open end 8 of the double-walled pipe 7 inside the cooling zone. The double-walled pipe 7 is fixed to the wall of the cooling zone by means of the flange 9 comprising the bores 10. The inert gas is introduced

through the feed pipe 11 into the space 12 between the inner wall 13 and the outer wall 14 of the double-walled pipe 7. The inert gas flows through the open end 13 of the space 12 into the interior of the cooling zone.

70 *Figure 3* shows the inner wall 13 and the outer wall 14 of the double-walled pipe joined together by the webs 16.

75 CLAIMS

1. A process for the pyrogenic production of a very finely divided oxide of a metal and/or metalloid by reacting a volatile halogen compound of the metal and/or metalloid with an oxyhydrogen gas mixture in which elementary halogen formed during formation of the oxide of the metal and/or metalloid is reacted during cooling of the reaction products in a cooling zone with additionally introduced gaseous hydrogen at a temperature below the reaction temperature of the gaseous hydrogen additionally introduced and the oxygen present in the waste reaction gases, to form hydrogen halide, the gaseous hydrogen being introduced into the cooling zone by means of at least one double-walled pipe, and an inert gas being additionally introduced by means of the same double-walled pipe between the outer wall and the inner wall of the said double-walled pipe.

95 2. A process as claimed in claim 1 wherein the hydrogen is introduced at a temperature of from 500 to 700°C.

3. A process as claimed in claim 2 wherein the hydrogen is introduced at a temperature of from 550 to 630°C.

100 4. A process as claimed in any of claims 1 to 3 wherein the inert gas is nitrogen.

5. A process as claimed in any of claims 1 to 3 wherein the inert gas is used in a quantity of from 20 to 200 m³/h.

6. A process as claimed in claim 5 wherein the inert gas is used in a quantity of from 50 to 150 m³/h.

7. A process as claimed in any of claims 1 to 6 wherein the volatile halogen compound is the corresponding chloride of the metal and/or metalloid.

8. A process as claimed in claim 1 substantially as described with particular reference to the accompanying drawings.

115 9. An oxide of a metal and/or metalloid when produced by a process as claimed in any of claims 1 to 8.